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RESEARCH ARTICLE

# Two halogenated flame retardants and cadmium in the soil-rice system: sorption, root uptake, and translocation

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## Abstract

The migration and transformation of Tetrabromobisphenol A (TBBPA), DechloranePlus (DP), and cadmium in soil-rice system was investigated, and the influence on the quality of two varieties of rice was studied. The degradation half-lives of TBBPA, BBPAs, syn-DP, and anti-DP were 23.18 ~ 26.36 days, 30.14 ~ 36.10 days, 72.96–81.55 days, and 169.06–198.04 days in the soil. TBBPA was gradually degraded to tri-BBPA, di-BBPA, mono-BBPA, and bisphenol A by the debromination. TBBPA and its bromide metabolites could be bioaccumulated in different tissues of rice; mono-BBPA and bisphenol A was easy to accumulate in the stems, and bisphenol A was easy to bioaccumulate in the grain. Comparing with single and compound pollution, there was no significant difference in bioaccumulation factors of two rice species. The grain of NO7 had stronger bioaccumulation ability to mono-BBPA and BPA than NO1, and there is no significant difference in TBBPA. Residual level of DP in the rice: roots > stems > grain; there was no significant difference in bioaccumulation of two varieties of rice. Cadmium was easily bioaccumulated in the roots of rice and translocated to the rice stems and grains. NO7 rice had stronger bioaccumulation and transport capacity than NO1. The effects of the three pollutants on the quality of two varieties of rice varied significantly; cadmium had the greatest effect on the iodine blue value (BV) and amylase activity of the grain. This study proved that selecting rice varieties with low bioaccumulation to polluters can effectively reduce the risk of the food chain harming human health.

**Keywords** Bioaccumulation · Tetrabromobisphenol A · Bisphenol A · Dechlorane plus · Soil-rice system · Migration and transformation

## Introduction

Dechlorane plus (DP) and Tetrabromobisphenol A (TBBPA) are halogenated flame retardants (HFRs) which have attracted more attention in recent years (Xian et al. 2011; Sun et al. 2014), and they have the characteristics of persistent organic pollutants (POPs). DP is a chlorinated flame retardants (CFRs) widely used in wire and cable coating structure, household appliances, and roofing plastic materials (Wang et al. 2010). TBBPA is widely used as brominated flame retardants (BFRs), and it is mainly used in resins as reactive flame retardants (FR) in the production of printed circuit boards (PCBS) (Wit 2002). Because DP has the characteristics of persistence, bioaccumulation, long-distance transmission, and so on, coupled with their widespread use, they are being detected in an increasing range of environments (Yin et al. 2020; Sverko et al. 2011). It is identified as a high production volume chemical by the US Environmental Protection Agency (2011), European Chemicals Agencies classified DP into Candidate List of Substances of Very High Concern. E-waste recycling areas are still the main source of DP pollution, which is especially evident in the southern part of the country (Ji et al. 2018).

TBBPA, poly brominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDs) are listed by the Stockholm Convention as persistent organic pollutants (POPs), the output of TBBPA accounts for about 50% of the

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total BFRs production, and it is the largest flame retardant product in the world, with an annual demand of over 170,000 t (Mäkinen et al. 2009). TBBPA has high log $K_{ow}$ (4.50 ~ 6.53) and BAF (bioaccumulation factors, 9.56 ~ 22.64), low water-soluble and high fat soluble. TBBPA has certain bioaccumulation effects (Environment Canada 2004), and it is easy to bio concentrate into organisms and produce toxicity on organisms. It has persistence and longer degradation half-life in the water and soil, and it stabilizes in various media such as water, soil and sediment. China has become the most polluted areas with TBBPA, the major production areas (Tianjin, Shandong, and Jiangsu) and electronic waste recycling areas (Zhejiang and Guangdong) are typical pollution areas (Johnson-Restrepo et al. 2008; Liu et al. 2016). The content of TBBPA in the sediment of wastewater is the highest, up to 41,200 ng/g (Li et al. 2019), which shows that TBBPA content is related to pollution sources. The concentration of TBBPA is 7758 ng/g and 646.04 ng/g in BFRs production area of Shouguang in Shandong province and electronic waste recycling area of Qingyuan in Guangdong (Zhu et al. 2014; Huang et al. 2014), respectively, which is significantly higher than unpolluted soil (5.6 ng/g) (Xu et al. 2012).

Cd is a kind of heavy metal substance; it has bioaccumulation, persistence, and irreversibility, and it has high toxicity and concealment. It is the "prime culprit" of bone pain. In recent years, a large number of researchers have carried out in-depth studies on the heavy metal pollution status in China. Nevertheless, cadmium residues in the soil ranged 0.211–14.9 mg/kg in the soil of Hunan province, accounting for 3.29 to 53.9% of the total heavy metal. The soil–plant system consists of inorganic parts of soil, organic parts and green higher plants, or crops, which are the basic structural units of the biosphere, the link between urban and rural ecosystems, and the bridge between plants and animals. Nowadays, a variety of studies have been conducted on the behavior of transport transformations of various pollutants in the soil–plant system and how to reduce pollution (Rajput et al. 2020; Knight et al. 2021). Cheng et al. (2020) studied the transformation and dissipation mechanisms of DP in the rhizosphere of the soil–plant system, and the depuration, accumulation, and translocation of DP in rice plants were observed. There are also studies on the natural attenuation and metabolism of TBBPA, and the migration and transformation of Cd were studied by means of isotope determination (Wang et al. 2021; Wiggenhauser et al. 2018; Imseng et al. 2018), but there are few studies on the mechanism.

Using the outdoor pot experiment method, the uptake, migration, and transformation of two halogenated flame retardants and Cd have been researched in the soil-rice system in the research. The effects of soil pollution on the quality of rice have been studied by measuring the water-soluble protein content, free fatty acids, BV, lipase activity, protease activity, α-amylase, and amylase in the two rice grains. It provides scientific basis for correctly evaluating the toxicity of flame retardants and Cd in soil to crops, and rice varieties insensitive to the pollutants were screened. The mechanisms of soil translocation and rice uptake of halogenated flame retardants and cadmium were measured, it provides scientific basis for the control technology and mechanism of pollutants. This study proves that selecting rice varieties with low bioaccumulation to pollutants can effectively reduce the risk of food chain harming human health.

## Materials and methods

### Experimental reagents

DP (Accu. Standard Inc., 100%), 50 µg/mL syn-DP and anti-DP standard sample (Accu Standard Inc., toluene solvent), N-hexane and dichloromethane (chromatographic purity), Tetrabromobisphenol A (Dr, German, 99.00%), Methanol and acetonitrile (chromatographic purity), 50 µg/mL Tetrabromobisphenol A standard sample (Methanol, Dr, German), and $CdCl_2 \cdot H_20$ (99.99%, Sigma-Aldrich).

Guosetianxiang NO1 (NO1) and Guosetianxiang NO7 (NO7), two kinds of rice seeds, are purchased in the market.

### Experimental method

The experiment was conducted at the experimental station of Shandong Agricultural University, which is located in Taian (Shandong Province, China). The rice plants were grown in plastic pots which contained 8 kg of soil; each plastic pot was placed on a plastic tray, passing air and preventing local contamination. The surface soil (0 ~ 20 cm) was collected from the experimental station of Shandong Agricultural University (the concentration of polluters has been determined in advance), and the tested soil was brown soil. The physical and chemical properties of the soil were as follows: organic nitrogen, available phosphorus, available potassium, and organic matter were 133.3, 18.6, 123.7 mg/kg, and 17.8 mg/kg, respectively, DP and TBBPA were not detected, and the local value of Cd was 0.25 mg/kg. According to the conventional fertilization method, adding 2% (weight ratio) fermented cow dung to the soil.

For the two rice species, five treatment groups were set up, which were CK, 6 mg/kg TBBPA, 5 mg/kg DP, 25 mg/kg Cd, and combined pollution (T + DP + Cd). In addition to the 10 parallels for combined pollution, five parallels were set up for each treatment group, with a total of 60 pots. Rice seed was grown after soil was balanced for 10 days, and the soil was fertilized and watered at different periods according to the rice growth.

The germination rate was measured after 5 days of planting and the aboveground part was measured as seedling length after 10 days of cultivation.

### Extraction and determination of two halogenated flame retardants and Cd in the samples

Samples were extracted by ASE, and TBBPA and its metabolites were determined by UltiMate-TSQ HPLC–MS/MS (Thermo Fisher Scientific) (Chang 2021). The residue amount of DP in the soil, root, and rice shoot and grain samples was detected by TSQ 8000 GC–MS/MS (Thermo Fisher Scientific) according to the literature (Xie et al. 2020). Residual level of Cd in rice and soil samples were determined by iCAP™ TQs ICP-MS (Thermo Fisher Scientific) (Jiang et al. 2020).

### Degraded dynamics of two halogenated flame retardants and Cd in soil

Soil samples were collected at 0, 20, 50, 80, and 140 days of rice planting. By detecting the concentration of two flame retardants and Cd in the soil at different times, the degradation and metabolites in the soil were studied, and exploring the degradation mechanism under the action of single and combined pollution.

### Determination of rice quality indicators

On the 140th days, the rice was harvested, peeled, and crushed, and quality indicators of rice were measured. The protein of grain was determined by national standards (GB-T5511-2008), crude fat content was detected by the Soxhlet Extractor Method ((NY/T 4–1982), the BV of the rice was determined by the iodine reagent method (Zhao et al. 2007). The activity of lipase of rice was determined by Lipase (LPS) Activity Assay Kit; the activity of protease was detected by Protease Activity Assay Kit; the activity of amylase and α-amylase was detected by the Soluble Starch Synthase (SSS) Activity Assay Kit and α-Amylase (α-AL) Activity Assay Kit. All assay kits were from *boxbio* (Beijing box Shenggong Technology Co., Ltd).

### Data statistics and analysis

The translocation behaviors of the TBBPA, DP, and Cd in the soil-rice system were investigated through bioaccumulation factor (BF) and translocation factor (TF). BF is the accumulation factor of pollutant from soil to root, $TF_{r\text{-}s}$ indicates the transformation coefficient of pollutant from roots to straw and $Tf_{s\text{-}g}$ is the conversion coefficient from straw to grain.

Residual concentration of polluters is presented as $\overline{x} \pm$ sd, the mean values compared using the Statistical Package for Social Sciences package (SPSS 22.0 for Windows) by a multiple comparison test at the 5% probability level. Figures were plotted by Sigma plot 12.5, and ANOVA was used to evaluate the significance by the least significant difference (LSD) ($p < 0.05$).

## Results and discussion

### Stress effect of two halogenated flame retardants and cadmium on rice germination and seedling growth

According to Table 1, the response of the two kinds of rice is different to pollutants. NO1 rice was more tolerant of cadmium pollution, there were no significant differences in germination percentage between 25 mg/kg treatment and the control, and there were significant differences between the other treatments and the control. NO7 rice could tolerate DP pollution, there were no significant differences in germination percentage between 5 mg/kg treatment and the control, and there were significant differences between the other treatments and the control. Rothenbacher and Pecquet (2018) reported the effect of TBBPA on the seedlings of six plants; the most sensitive endpoints were dry weight and height of seedling; different plants had different sensitivity to TBBPA; cucumber was sensitive (20 mg/kg); and soybean had the best tolerance to TBBPA.

**Table 1** The stress response of the two kinds of rice to pollutants in the different treatment

| Rice type | Indicators | Treatment | | | | |
|---|---|---|---|---|---|---|
| | | CK | DP | TBBPA | Cd | Combined pollution |
| NO1 | Germination rate (%) | 80.25±5.15a | 65.08±3.56c | 75.06±3.21b | 80.00±4.21a | 62.25±3.15c |
| | Height(cm) | 5.85±0.25a | 4.32±0.20c | 4.21±0.18c | 4.80±0.12b | 3.92±0.26d |
| NO7 | Germination rate (%) | 86.58±5.02a | 77.89±4.15a | 59.58±4.02b | 52.28±4.02bc | 46.53±2.16c |
| | Height(cm) | 5.25±0.18a | 3.08±0.23d | 4.10±0.20b | 3.42±0.12c | 2.98±0.13d |

mean value is expressed as $\overline{x} \pm$ sd ; different letters (a, b, c, and d) mean significant differences in the comparison among different treatments ($p < 0.05$); on the contrary, having no significant differences ($p > 0.05$)

Pollutants could adversely affect seedling germination and growth. In the treatment of DP, TBBPA, Cd, and combined pollution, the inhibiting rates of NO1 rice height were 26.15%, 28.03%, 17.95, and 32.99%, and the inhibiting rates of NO7 rice height were 41.33%, 21.90%, 34.86, and 43.24%, respectively. It indicated that the germination and seedling growth of rice were greatly affected by the pollutants; the result was consistent with the literature (Ge and Zhang 2017). Ge and Zhang (2017) reported that TBBPA (5 ~ 100 mg/kg) significantly inhibited the growth of soybean seedlings. With the increase in TBBPA concentration, the toxic effect was greater. Wang et al. (2015) reported that rice seedlings produced more MDA and higher activities of antioxidant enzymes under the stress of cadmium. This study showed that the response of two types of rice to cadmium was significantly different; the tolerance of NO1 was more than NO7 to cadmium.

## Migration and transformation of halogenated flame retardants and cadmium

By detecting the concentration of TBBPA in soil with different treatments, its degradation and metabolites in soil were studied, and the degradation dynamics of TBBPA in soil with two types of rice are shown in Fig. 1, and the parameters of TBBPA and BBPAs in the Exponential Decay degradation model are shown in Table 2. The degradation half-lives of TBBPA were 23.18 ~ 26.36 days in the soil with two rice varieties, and BBPAs were 30.14 ~ 36.10 days, and there was no significant difference between the two kinds of rice. The residual amount decreased by 94.42% and 96.37% in the soil with NO1 rice and by 90.57% and 95.62% in the soil with NO7 rice during the 140-day trial period, respectively. In the single pollution treatment, TBBPA and BBPAs in soil were degraded rapidly in the first 50 days; the concentration of TBBPA reduced from 6682.52 to 1244.41 ng/g and 1270.32 ng/g in the soil with NO1 rice and NO7 rice, respectively; in the combined pollution, the concentration of TBBPA reduced from 5576.13 to 1150.05 ng/g and 1200.72 ng/g in the soil with NO1 rice and NO7 rice, there was no significant difference in the degradation rate of TBBPA in the soil. The degradation rate was slowed down after 50 days; the residual concentrations of TBBPA were 372.51 ng/g and 630.31 ng/g in the soil with single pollution on 140 days, and the concentrations were 202.21 and 244.14 ng/g in the soil with combined pollution. The residual concentrations of TBBPA varied significantly in the soil with two kinds of rice, the degradation of TBBPA was significantly different in the soils with two kinds of rice.

Sun et al. (2014) reported that TBBPA dissipation (half-life 20.8 days) was accompanied by mineralization (11.5% of initial TBBPA) in unplanted soil, and TBBPA dissipated with the degradation half-life of 14.7 days (Li et al. 2015). Wei et al. (2018) reported that the dehalogenation of TBBPA was likely a stepwise removal of bromine atoms; the pathway of TBBPA → tri-BBPA → di-BBPA → mono-BBPA → BPA was thus proposed for TBBPA degradation, and the microbial activity was the critical factor for the degradation of TBBPA; the degradation of TBBPA in plant-soil systems was biologically mediated. Monoculture reed growth and rice–wheat rotation could accelerate TBBPA and BPA removal in the surface soil layers by stimulating the anaerobic debromination in the rhizosphere soil, but the decrease in the concentration of TBBPA in the deep soil layers was not impacted by rice–wheat rotation or reed planting (Wang et al. 2021). Anaerobic degradation of TBBPA in river sediment was enhanced with the addition of humic acid, sodium chloride, zero-valentiron, vitamin B12, rhamnolipid, and surfactin, but it was inhibited by the addition of acetate, lactate, and pyruvate (Chang et al. 2012). In this study, two kinds of rice were planted in the polluted soil. The rice species had certain effect on the degradation rate of TBBPA in the soil, and BPA and mono-BBPA were the main metabolites. The degradation half-life was longer; it was possible that microbial activity in the soil was different.

By detecting the concentration of syn-DP and anti-DP in the soil, its dynamic changes in the soil were studied, and the degradation mechanism in the alone and combined pollution has been explored (Fig. 1). Parameters of syn-DP, anti-DP, and DP in the Exponential Decay degradation model are shown in Table 3. The results showed that degradation half-lives of syn-DP and anti-DP were 72.96–81.55 days and 169.06–198.04 days, different treatments had little effect, and there was no significant difference in degradation half-life between the two rice varieties.

During the 140-day test period, syn-DP was reduced by 71.54% and 74.57% in the soil of single and combined pollution with NO1 rice, and the anti-DP decreased by 43.03% and 43.47%. The concentration of DP in the soil with DP treatment was reduced from 5.25 to 2.64 μg/g, and DP in the combined pollution was reduced from 5.16 to 2.55 μg/g. In the soil of single and combined pollution with NO7 rice, syn-DP was reduced by 73.98% and 75.42%, and anti-DP was reduced by 45.27% and 45.23%. The concentration of DP in the soil with single pollution was reduced from 5.25 to 2.52 μg/g, and DP with combined pollution was reduced from 5.16 to 2.47 μg/g. There was no significant difference in the residual concentration in the soil between single and combined contamination in the same period ($p > 0.05$), and there was no significant difference between the two rice species ($p > 0.05$).

Tao (2020) summarized the degradation of DP in soil–plant systems by four main pathways: dechlorination, oxidation, cleavage, and isomerization; its migration and transformation mechanisms were investigated. Sun et al. (2019) reported that the half-lives of DP dissipation in soil were 70 ~ 102 days, and the dissipation of DP in greenhouse soil was slightly slower than that in conventional soil. In this study, the degradation

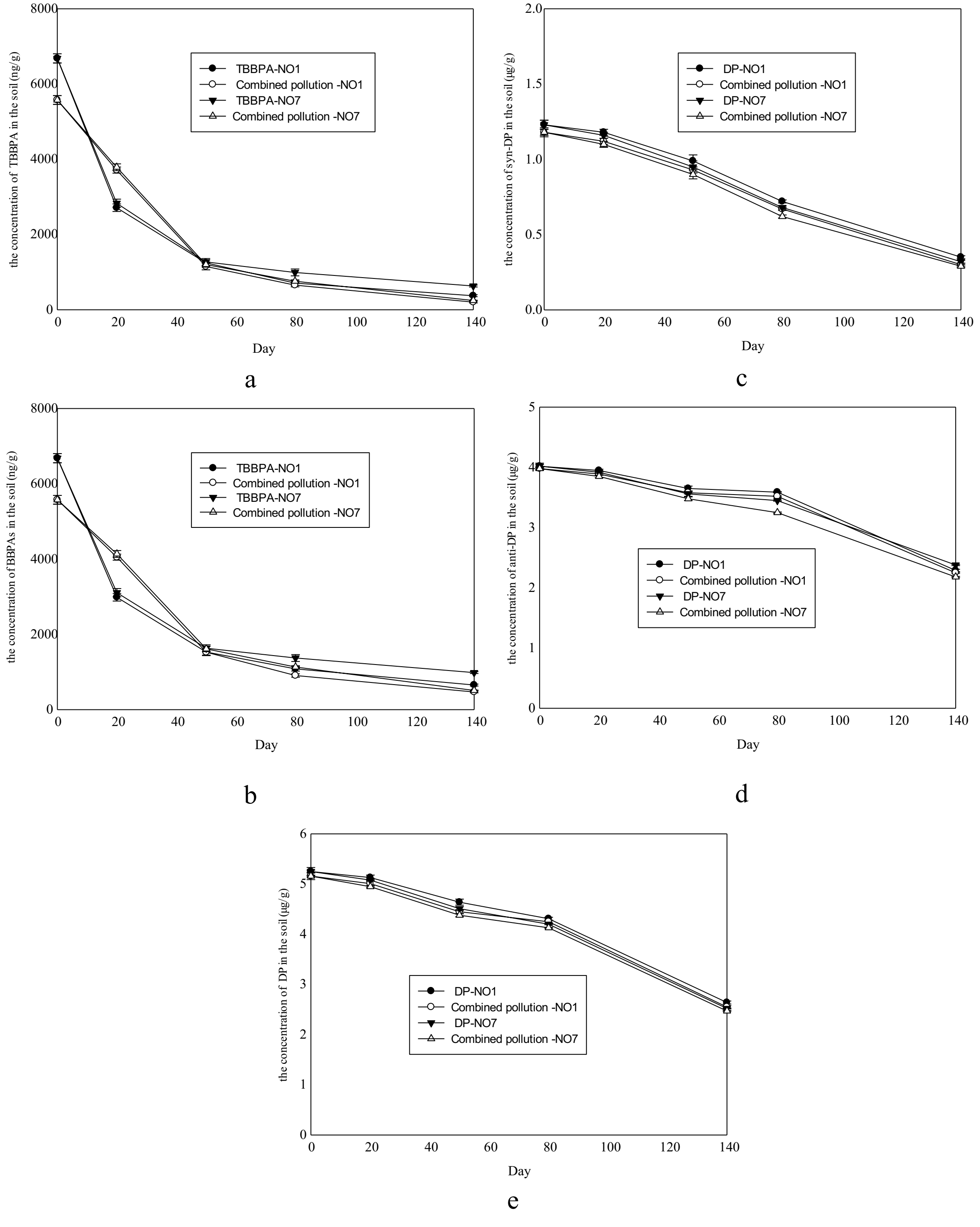


**Fig. 1** Degradation dynamics of two halogenated flame retardants in the soil with two types of rice. Note: Figures a–e represent the degradation dynamics of TBBPA, BBPAs, syn-DP, anti-DP, and total DP in the different treatments, respectively

half-lives of DP in soil were 141.46 ~ 150.68 days. The degradation half-life is longer than that reported and may be caused by differences in soil properties. There was no significant difference in the degradation rate of DP between the soils with two kinds of rice; the removal rate of syn-DP is 71.54 ~ 75.42% and anti-DP is 43.03 ~ 45.27% (Fig. 1).

The concentration of Cd in the soil is shown in Table 4. The study showed that within the 140 days of the test, cadmium decreased by 26.12% (single pollution) and 20.61% (combined pollution) in the soils with NO1 rice, Cd reduced by 28.95% (single pollution) in the 21.94% (combined pollution) in the soils with NO7 rice. Because heavy metals

**Table 2** Degradation parameters of TBBPA and BBPAs in the soil by the exponential decay degradation model

| Treatment | Pollutants | degradation model $C = a\cdot e^{-bt}$ | *P*(a) | *P*(b) | r | $T_{1/2}$(d) |
|---|---|---|---|---|---|---|
| TBBPA-NO1 | TBBPA | $C = 5987.1305e^{-0.0299t}$ | 0.0065 | 0.0116 | 0.9675 | 23.18 |
| Combined pollution -NO1 |  | $C = 5698.6244e^{-0.0277t}$ | 0.0013 | 0.0021 | 0.9902 | 25.02 |
| TBBPA-NO7 |  | $C = 5810.7187e^{-0.0264t}$ | 0.0140 | 0.0269 | 0.9342 | 26.25 |
| Combined pollution -NO7 |  | $C = 5666.1299e^{-0.0262t}$ | 0.0017 | 0.0028 | 0.9880 | 26.46 |
| TBBPA-NO1 | BBPAs | $C = 5730.5414e^{-0.0230t}$ | 0.0102 | 0.0202 | 0.9462 | 30.14 |
| Combined pollution -NO1 |  | $C = 5631.1095e^{-0.0222t}$ | 0.0021 | 0.0040 | 0.9833 | 31.22 |
| TBBPA-NO7 |  | $C = 5498.4783e^{-0.0192t}$ | 0.0167 | 0.0404 | 0.9063 | 36.10 |
| Combined pollution -NO7 |  | $C = 5575.7580e^{-0.0202t}$ | 0.0019 | 0.0040 | 0.9836 | 34.31 |

*P*(a) and *P*(b) < 0.05, it indicates that there is significant differences. $T_{1/2}$(d) is degradation half-life of pollutants

**Table 3** Degradation parameters of DP in the soil by the exponential decay degradation model

| Pollutants | Treatment | degradation model $C = a\cdot e^{-bt}$ | *P*(a) | *P*(b) | r | $T_{1/2}$(d) |
|---|---|---|---|---|---|---|
| Syn-DP | DP-NO1 | $C = 1.3418e^{-0.0085t}$ | 0.008 | 0.0057 | 0.9782 | 81.55 |
|  | Combined pollution -NO1 | $C = 1.2910e^{-0.0092t}$ | 0.001 | 0.006 | 0.9777 | 75.34 |
|  | DP-NO7 | $C = 1.3352e^{-0.0091t}$ | 0.0007 | 0.0043 | 0.9821 | 76.17 |
|  | Combined pollution -NO7 | $C = 1.2784e^{-0.0095t}$ | 0.0007 | 0.0039 | 0.9834 | 72.96 |
| Anti-DP | DP-NO1 | $C = 4.2495e^{-0.0037t}$ | 0.0005 | 0.0247 | 0.9340 | 187.33 |
|  | Combined pollution -NO1 | $C = 4.1934e^{-0.0038t}$ | 0.0004 | 0.0220 | 0.9389 | 182.41 |
|  | DP-NO7 | $C = 4.1843e^{-0.0035t}$ | 0.002 | 0.0107 | 0.9618 | 198.04 |
|  | Combined pollution -NO1 | $C = 4.1537e^{-0.0041t}$ | 0.002 | 0.0070 | 0.9717 | 169.06 |
| DP | DP-NO1 | $C = 5.575e^{-0.0046t}$ | 0.0004 | 0.0129 | 0.9585 | 150.68 |
|  | Combined pollution -NO1 | $C = 5.4553e^{-0.0047t}$ | 0.0005 | 0.0131 | 0.9579 | 147.48 |
|  | DP-NO7 | $C = 5.5519e^{-0.0049t}$ | 0.0004 | 0.0103 | 0.9646 | 141.46 |
|  | Combined pollution -NO1 | $C = 5.42996e^{-0.0049t}$ | 0.0004 | 0.0101 | 0.9650 | 141.46 |

*P*(a) and *P*(b) < 0.05, it indicated that there was significant differences. $T_{1/2}$(d) is degradation half-life of pollutants

**Table 4** Residual concentration of cadmium at different times in the soil with two types of rice

| Time (day) | Treatment |  |  |  |
|---|---|---|---|---|
|  | Cd-NO1 | Combined pollution-NO1 | Cd-NO7 | Combined pollution-NO7 |
| 0 | 23.66 ± 0.71 | 21.88 ± 0.72 | 23.66 ± 0.51 | 21.88 ± 0.62 |
| 20 | 22.08 ± 0.82 | 20.68 ± 0.72 | 23.23 ± 0.80 | 20.72 ± 0.83 |
| 50 | 19.58 ± 0.87 | 20.05 ± 0.53 | 19.48 ± 0.73 | 20.59 ± 0.75 |
| 80 | 19.89 ± 0.56 | 19.14 ± 0.34 | 19.28 ± 0.64 | 20.06 ± 0.56 |
| 140 | 17.48 ± 0.62 | 17.37 ± 0.69 | 16.81 ± 0.59 | 17.08 ± 0.65 |

Residual concentration of Cd is indicated as $\overline{x} \pm sd$, and the unit is mg/kg

were difficult to degrade in the soil, the disappeared cadmium in the soil was probably absorbed by rice; cadmium residue in the grain, root, and leaves of rice illustrated this issue. As a nonessential element, Cd was potentially assimilated by plants (Riaz et al. 2021). Because of its high toxicity and widespread pollution, Cd contamination in paddy fields was a serious health concern (Li et al. 2021). In this study, Cd could be absorbed by rice root and entered the plant for migration and transformation.

## Rice sorption and root uptake for two halogenated flame retardants and cadmium

Two types of rice could absorb two halogenic flame retardants and cadmium in the soil, and the residual levels of TBBPA, DP, and Cd in different tissues of rice are summarized in Tables 5 and 6. The accumulation regularity of two halogenic flame retardants and Cd was explored in two types of rice and its tissues. The risk of biological amplification by absorbing flame retardants and Cd into the food chain was studied, and rice varieties with low bioaccumulation for pollutants were selected in order to reduce the risk of endangering human health by the food chain.

TBBPA and its bromide metabolites were bioaccumulated in different rice tissue sites. The residual amount of TBBPA and its metabolites such as di-BBPA, mono-BBPA, and bisphenol A in the root of NO1 rice was more than tri-BBPA; the amount of mono-BBPA and bisphenol A in the shoot of rice was higher; bisphenol A in the grain of rice was higher, it was largely related to the characteristic of the metabolites in the metabolism of TBBPA, and TBBPA and tri-BBPA were easily debromated to generate new substances, while BPA was relatively stable and it degraded slowly. In the treatment of TBBPA, BFs of TBBPA, mono-BBPA, and bisphenol A were 0.0713, 1.719, and 1.792; $TF_{r\text{-}s}$ was 0.116, 0.363, and 0.179; TFs-g was 1.080, 0.277, and 0.535. In the treatment of combined pollution, BFs of TBBPA, mono-BBPA, and bisphenol A were 0.0451, 1.744, and 0.771; TFr-s was 0.142, 0.368, and 0.402; TFs-g was 0.606, 0.541, and 0.653. There was no significant difference between the two treatments in bioaccumulation and bio transfer capacity.

For the NO7 rice, the amount of TBBPA and bisphenol A in the root was higher, while the amount of tri-BBPA was relatively less; the amount of mono-BBPA and bisphenol A in the shoot was relatively higher; the amount of bisphenol A in the grain of rice was higher; there were significant differences between the two treatments ($p < 0.05$); and there were significant differences in the residual levels of TBBPA and BPA between the two rice species ($p < 0.05$); it indicated that the type of rice affected its amount of absorbing pollutants, and the variety of rice determined the extent of absorbing pollutants, and it provided the basis for the selection of planting rice varieties. In the TBBPA treatment, BFs of

**Table 5** Residual concentration of TBBPA and its bromine metabolites in the tissue of two kinds of rice

| Rice type | Tissue | Treatment | TBBPA and its bromine metabolites | | | | |
|---|---|---|---|---|---|---|---|
| | | | TBBPA | Tri-BBPA | Di-BBPA | Mono-BBPA | BPA |
| NO1 | root | TBBPA | 193.22 ± 17.12a | 14.13 ± 0.52a | 184.25 ± 8.72a | 195.92 ± 3.72a | 321.51 ± 5.72a |
| | | Combined pollution | 167.61 ± 15.63b | 14.82 ± 0.32a | 101.82 ± 2.32b | 197.42 ± 2.72a | 136.51 ± 1.72b |
| | straw | TBBPA | 22.32 ± 2.55a | 25.3 ± 0.76a | 43.20 ± 0.72a | 71.12 ± 2.43a | 57.82 ± 0.30a |
| | | Combined pollution | 23.72 ± 0.95a | 8.72 ± 0.22b | 40.75 ± 0.58b | 72.71 ± 3.47a | 54.85 ± 0.64b |
| | grains | TBBPA | 24.12 ± 0.85a | 4.02 ± 0.72a | 21.61 ± 0.34b | 19.72 ± 0.62b | 30.92 ± 0.72b |
| | | Combination | 14.37 ± 0.72b | 1.52 ± 0.07b | 33.32 ± 0.28a | 39.32 ± 0.72a | 35.81 ± 0.91a |
| NO7 | root | TBBPA | 219.71 ± 15.32a | 11.48 ± 0.09a | 65.54 ± 0.65a | 87.04 ± 0.35a | 140.99 ± 1.85a |
| | | Combined pollution | 83.11 ± 1.12b | 7.98 ± 0.05b | 49.24 ± 0.85b | 79.56 ± 0.25b | 124.24 ± 2.05b |
| | straw | TBBPA | 26.38 ± 0.85a | 19.12 ± 0.15a | 57.01 ± 0.56a | 67.82 ± 0.15b | 69.78 ± 0.95a |
| | | Combined pollution | 25.56 ± 0.65a | 12.25 ± 0.25b | 36.74 ± 0.76b | 72.83 ± 0.25a | 65.64 ± 0.89b |
| | grains | TBBPA | 12.87 ± 0.25a | 1.66 ± 0.05b | 19.40 ± 0.15a | 19.17 ± 0.15b | 36.30 ± 0.55b |
| | | Combined pollution | 13.09 ± 0.21a | 2.73 ± 0.04a | 20.74 ± 0.10a | 57.33 ± 0.10a | 63.91 ± 0.98a |

The mean value is expressed as $\bar{x}$ ± sd, the unit is ng/g; different letters (a and b) mean significant differences in the comparison between different treatments ($p < 0.05$); on the contrary, having no significant differences ($p > 0.05$)

**Table 6** Residual concentration of DP and Cd in the tissue of two kinds of rice

| Treatment | Pollutants | NO1 rice | | | NO7 rice | | |
|---|---|---|---|---|---|---|---|
| | | Root | Straw | Grain | Root | Straw | Grain |
| DP | Syn-DP | 22.58 ± 1.72a | 5.46 ± 0.29b | 1.29 ± 0.03c | 20.24 ± 0.24a | 4.89 ± 0.25b | 1.08 ± 0.02c |
| | Anti-DP | 55.18 ± 2.01a | 15.25 ± 0.25b | 3.56 ± 0.02c | 53.18 ± 0.42a | 13.29 ± 0.13b | 3.66 ± 0.07c |
| Combined pollution | Syn-DP | 20.19 ± 0.15a | 4.85 ± 0.12b | 1.78 ± 0.05c | 20.05 ± 0.18a | 4.85 ± 0.07b | 1.79 ± 0.02c |
| | Anti-DP | 52.67 ± 0.35a | 13.56 ± 0.28b | 3.83 ± 0.03c | 51.43 ± 0.16a | 12.65 ± 0.09b | 3.28 ± 0.03c |
| Cd | Cd | 71.17 ± 1.71a | 7.46 ± 0.38b | 1.29 ± 0.02c | 77.23 ± 0.25a | 14.13 ± 0.78b | 3.03 ± 0.04c |
| Combined pollution | Cd | 58.86 ± 2.56a | 6.81 ± 0.29b | 3.03 ± 0.09c | 80.77 ± 2.17a | 7.23 ± 0.48b | 4.78 ± 0.16c |

The mean value is expressed as $\bar{x}$ ± sd, the unit of DP is ng/g, the unit of Cd is μg/g; different letters (a, b, and c) mean significant differences ($p < 0.05$) in the comparison among parts of rice; on the contrary, having no significant differences ($p > 0.05$)

TBBPA, mono-BBPA, and bisphenol A were 0.078, 0.767, and 0.748; TFr-s were 0.120, 0.779, and 0.495; TFs-g were 0.488, 0.283, and 0.520. In the treatment of combined pollution, BFs of TBBPA, mono-BBPA, and bisphenol A were 0.022, 0.630, and 0.613; TFr-s were 0.308, 0.915, and 0.528; TFs-g were 0.512, 0.787, and 0.974, and there was no significant difference between the two treatments in bioaccumulation and bio transfer capacity. Grain of NO7 had stronger bioaccumulation ability to mono-BBPA and BPA than NO1, and there was no significant difference in TBBPA.

It was reported that crops could absorb TBBPA and its metabolites; lettuce and tomato could uptake BPA from soil and it be bio accumulated in the edible part; the higher amount of BPA in two rhizospheres may cause potential risk (Lu et al. 2015). Wang et al. (2016) reported that TBBPA could be absorbed in the rice cell suspension culture, the cells could accumulate TBBPA in the cytoplasm, the majority of the accumulated residues (70–79%) in the cells were attributed to the cellular debris-bound residues, and a small amount of DBHPA was detectable inside the cells. In this study, DBHPA was not detected in rice, but some bromine metabolites have been detected. Sun et al. (2014) proved that TBBPA dissipation was slightly accelerated in the rice soil; rice seedlings showed a high potential to accumulate TBBPA and its metabolites from the soil (21.3%); reed seedlings could accumulate significantly TBBPA and its metabolites. In addition, TBBPA in the contaminated sediment could be taken up by roots of mangrove species and then translocated to aboveground tissues (Jiang et al. 2020).

The residual levels of syn-DP and anti-DP in the rice with two treatments followed the sequence of roots > straw > grain, but there was no significant difference in the residual amount between the two rice tissues ($p > 0.05$). For NO1 rice, the BF of syn-DP was 0.0183 and 0.0171 in the single pollution and combined pollution treatment, TFr-s was 0.242 and 0.240, TFs-g was 0.236 and 0.367, the BF of anti-DP was 0.0137 and 0.0132 in the single pollution and combined pollution treatment, TFr-s was 0.276 and 0.257, and TFs-g was 0.233 and 0.260. For NO7 rice, the BF of syn-DP was 0.0165 and 0.0170 in the single and combined pollution treatment, TFr-s was 0.240 and 0.242, TFs-g was 0.223 and 0.369, the BF of anti-DP was 0.0132 and 0.0129 in the single pollution and combined pollution treatment, TFr-s was 0.255 and 0.246, and TFs-g was 0.270 and 0.259.

DP was selectively accumulated in plant tissues, and translocation factors and log $K_{ow}$ were positively correlated during translocation from root to stem. Cheng et al. (2020) reported that rice could uptake, translocate, depurate, and accumulate DP, rice plants tended to selectively absorb syn-DP from soil. Similar stereo selective bioaccumulation of syn-DP has been reported in eucalyptus foliage, pine needles, and lichens (Chen et al. 2011; Yang et al. 2016). Compared to anti-DP, syn-DP was the more lipophilic, and it was more prone to being trapped in root lipids and less likely to undergo translocation to aerial tissues (Cheng et al. 2020). In this study, it has been consistent with this research. In the treatment of high concentration, translocation played more important role in halogenated flame retardants burden than atmospheric uptake in leaf (Zhang et al. 2015). Rice, spinach, and tomato had potential uptake for DP, Zhang et al. (2015) reported that bioaccumulation factors were in the range of 0.32–2.2 for rice, 9.5–40 for spinach, and 1.6–7.7 for tomato. In this research, the bioaccumulation factors of rice root for DP were less compared with the plants reported in the literature.

The results showed that the residual concentration of Cd was high in the roots of the two types of rice; it was much more than residual level in the soil, which indicated that rice roots had a strong biological accumulation capacity for Cd. For the NO1 rice, BF was 4.07 and 3.39 in the single and combined pollution treatments; BF was 4.59 and 4.72 for NO7 rice. However, the residual concentrations in the straw and grain were lower than the root; $TF_{r\text{-}s}$ was 0.10 and 0.11 for the NO1, 0.18 and 0.09 for the NO7; $TF_{s\text{-}g}$ was 0.17 and 0.44 for the NO1, 0.21 and 0.66 for the NO7 rice. The two rice species with large differences of quality were selected for this study, and the residual levels of cadmium in the two rice species were significantly different ($p < 0.05$). NO7 rice had stronger bioaccumulation and transport capacity than NO1, which indicated that there was a significant difference in the absorption for cadmium between the two types of rice. Paddy rice was considered as main source for human exposure to Cd contamination due to its efficient accumulation especially when cultivated in contaminated fields. Du et al. (2018) reported that rice root was the main organ to take in Cd through chelation and adsorption, the translocation behaviors of Cd in the soil-rice system were investigated through bioaccumulation factor (BF) and translocation factor (TF). BF, $TF_{r\text{-}s}$, and $TF_{s\text{-}g}$ were 5.35, 0.18, and 0.41, respectively. Yang et al. (2021) reported that the BF of high-Cd-accumulating rice and the normal rice line were 2.3–4.6 and 1.58–3.14, TF was 0.09–0.16 and 0.02–0.09, respectively. In this study, BF was 4.39–4.72, $TF_{r\text{-}s}$ and $TF_{s\text{-}g}$ were 0.09–0.18 and 0.17–0.66, and it was consistent with the results of previous literature. Biochar could significantly reduce the accumulation of cadmium in rice, and biochar could be strategized in mitigating Cd-contamination in paddy soils and it could decrease Cd concentrations in rice (Rizwan et al. 2017).

## Effects of two halogenated flame retardants and Cd on two rice grain quality

By determining the water-soluble protein content, free fatty acid, BV, lipase activity, protease activity, α-amylase, and amylase of rice grain, the effect of pollutants on the quality of two rice had been investigated thoroughly, and it provided scientific basis for evaluating the toxicity of pollutants to crop. Two kinds of rice had different responses to pollutants and rice varieties insensitive to three pollutants could be selected (Fig. 2).

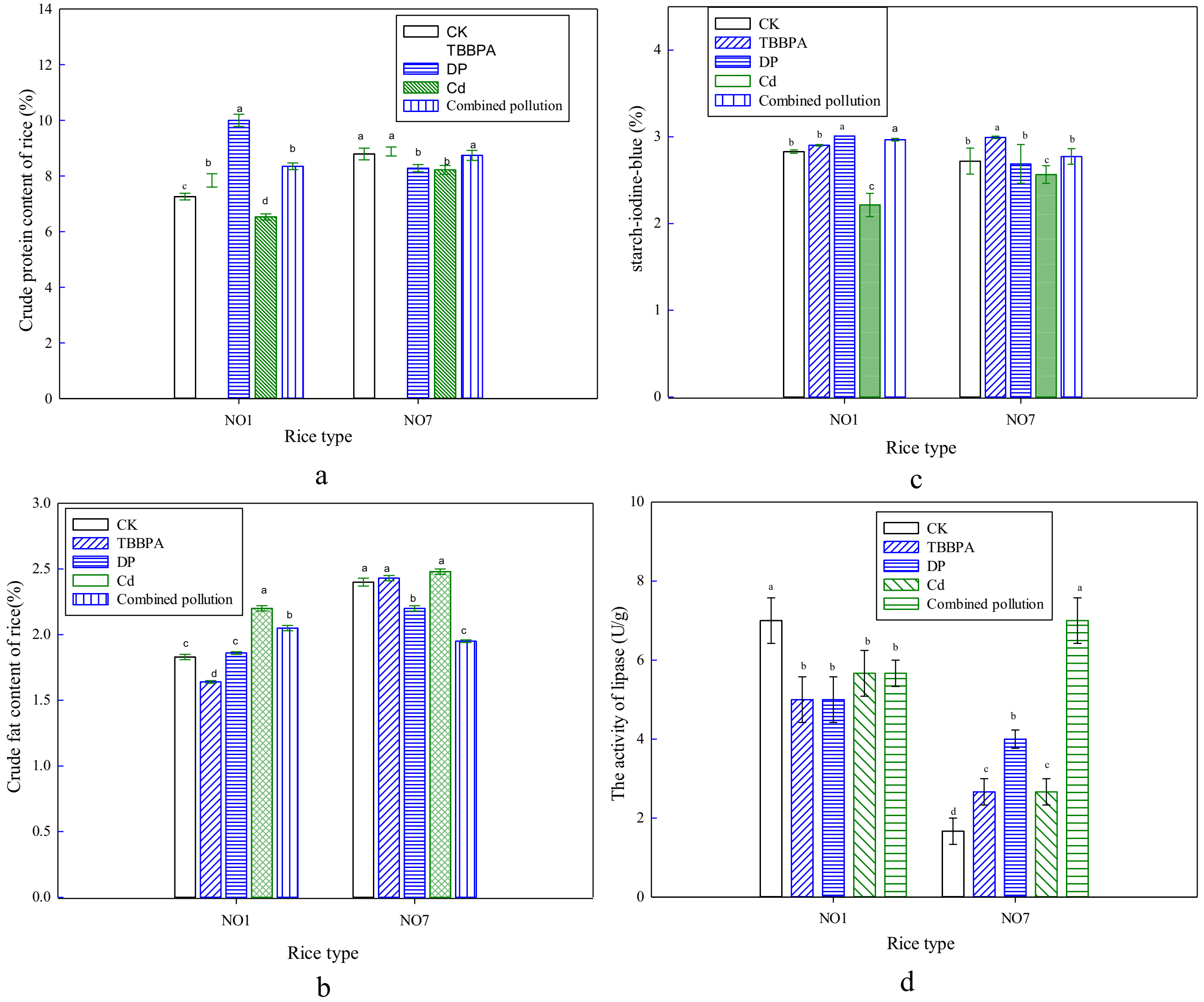
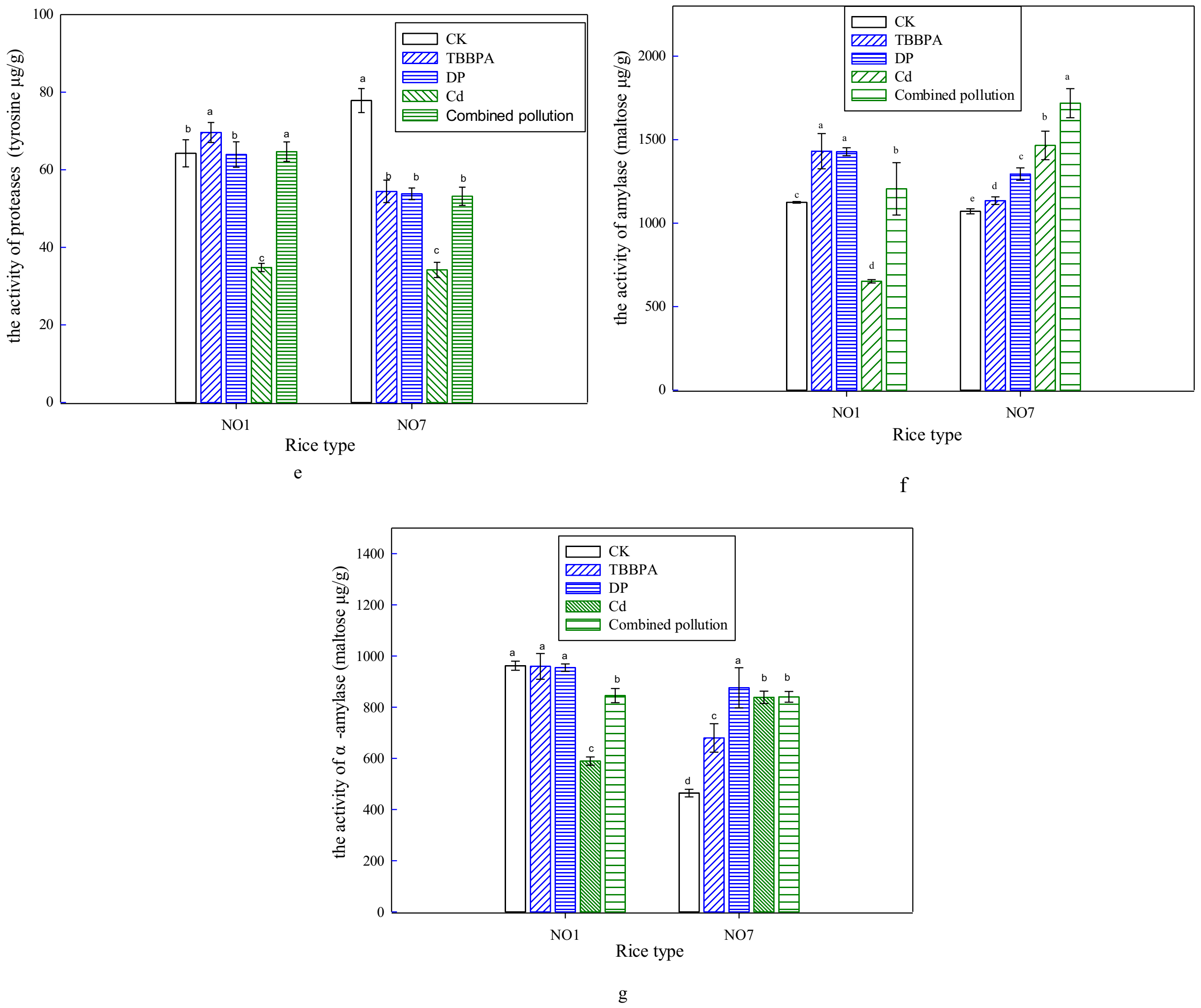


**Fig. 2** Effect of two halogenated flame retardants and Cd in soil on the quality of two types of rice. Note: Figures **a**–**g** represent the crude protein, crude fat, starch iodine blue value, lipase activity, protease activity, amylase activity and α- amylase activity of rice in the different treatments, respectively. Different letters (**a**, **b**, **c**, and **d**) mean significant differences in the comparison among different treatments ($p < 0.05$); on the contrary, having no significant differences ($p > 0.05$)

The protein content of NO1 rice increased by 7.99%, 37.74%, and 15.01% in the treatments of TBBPA, DP, and its combined contamination, but it was inhibited by 10.05% under the action of cadmium pollution. The response of NO7 to pollutants was different from NO1 rice; the protein content was slightly inhibited by 5.80% and 6.48% in the treatment of DP and Cd, there was no significant difference between the treatment of TBBPA, combined contamination, and the control ($p > 0.05$). After Cd ions enter plant cell, they form metal complexes or chelates with other compounds and thus inhibit metabolic processes (Yang et al. 2021), especially protein synthesis (Hall 2002). The content of soluble protein of rice was declined because of two reasons: Cd could promote proteolytic enzyme, resulting in proteolysis of existing protein; Cd could weaken synthesis of new proteins due to its toxicity effects on biosynthetic enzymes and organelles related to protein synthesis (John et al. 2009; Singh et al. 2006; Hall 2002).

For NO1 rice, the inhibition ratio of crude protein was 10.38% by TBBPA, there was no significant difference between the DP and the control. For NO7 rice, the inhibition ratio of crude protein was 8.33% and 18.75% in the treatment of DP and combined pollution, there were no significant differences between other treatments and controls ($p > 0.05$). The results showed that there were relatively significant differences in the responses of two varieties of rice to pollutants, and the mechanism needed further exploration.

Cadmium had an inhibiting effect on BV in two varieties of rice, and there were significant differences between Cd and



e

f

g

**Fig. 2** (continued)

other treatments ($p < 0.05$). The lipase activities of two varieties rice showed different responses to DP, TBBPA, and cadmium, the lipase activity of NO1 rice was inhibited, while the lipase activity of NO7 rice was significantly activated, which was significantly different compared with the control. The inhibition rate of cadmium on the protease activity of NO1 was 45.81%, and the protease activity was slightly promoted by TBBPA (8.37%), while there were no significant differences between DP, combined contamination, and the control. The protease activity of NO7 was inhibited by TBBPA, DP, cadmium, and combined pollution, and inhibition rates were 30.08%, 30.89, 56.10%, and 31.71%, respectively. For NO1 rice, the activity of amylase was promoted by TBBPA, DP and combined pollution, α-amylase activity was inhibited under the action of three pollutants, and the activity of α-amylase and amylase was inhibited by cadmium, inhibition rates were 38.77% and 42.08%, respectively. The response of two rice species to the pollutants was different, and the activities of α-amylase and amylase in NO7 rice were significantly improved under the action of the three pollutants.

## Conclusion

In the rice-soil system, two halogenated flame retardants and cadmium in the soil could be transferred to the food chain. Two kinds of rice had different responses to pollutants, and rice varieties insensitive to three pollutants could be selected. It proved that selecting rice varieties with low bioaccumulation to pollutants could effectively reduce the risk of food chain harming human health.

**Author contribution** Hui Xie: writing original draft and editing, supervision, data curation, resources, project administration, funding acquisition. Xin Liu: Writing—original draft, formal analysis, data curation, software. Yuxin Xu: investigation, resources, supervision. Ruiyuan Liu: conceptualization, methodology, formal analysis.

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**Data availability** Data, associated metadata, and calculation tools are available from the corresponding author (huixie@sdau.edu.cn).

## Declarations

**Ethical approval** This work does not contain any study with humans or animals.

**Consent to participate** Not applicable.

**Consent for publication** Not applicable.

**Competing interests** The authors declare no competing interests.

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